

Minimum requirements for laser-induced symmetry breaking in quantum and classical mechanics

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Abstract

Necessary conditions for generating phase controllable asymmetry in spatially symmetric systems using lasers are identified and are shown to be identical in quantum and classical mechanics. First, by studying the exact dynamics of harmonic systems in the presence of an arbitrary radiation field, it is demonstrated that anharmonicities in the system's potential are a necessary requirement for phase controllability. Then, by analyzing the space-time symmetries of the laser-driven Liouville dynamics for classical and quantum systems, a common set of temporal symmetries for the driving field that need to be violated to induce transport are identified. The conditions apply to continuous wave lasers and to symmetry breaking effects that do not rely on the control of the absolute phase of the field. Known examples of laser fields that can induce transport in symmetric systems are seen to be particular cases of these symmetry constraints.

I. INTRODUCTION

Recent years have witnessed the birth and rapid development of the coherent control field [1, 2, 3, 4, 5, 6], in which the coherence properties of applied laser fields are employed to steer a given quantum dynamical process in a desired direction. Of the different control schemes that have so far been developed, there is a general class that has the ability to induce phase controllable transport in spatially symmetric systems without introducing a bias voltage in the potential, a phenomenon that is referred to as laser-induced symmetry breaking.

This symmetry breaking effect is typically achieved by driving the system with AC fields with frequency components $n\omega$ and $m\omega$, where n and m are integers of different parity [1]. The nonlinear response of the system to such fields results in net dipoles or currents whose magnitude and sign can be manipulated by varying the relative phase between the two frequency components of the radiation [7]. For the popular case of $n = 1$ and $m = 2$ the rectification effect first appears in the third order response of the system to the incident radiation. At this order the system mixes the frequencies and harmonics of the field, generating a phase-controllable zero-harmonic (DC) component in the response.

Laser-induced symmetry breaking has been demonstrated in a wide variety of systems ranging from atoms to solid state samples. Experimentally it has been implemented for generating anisotropy in atomic photoionization [8], symmetry breaking effects in molecular photodissociation [9] (see also Ref. 10), photocurrents in quantum wells [11] and intrinsic semiconductors [12], as well as directed diffusion in symmetric optical lattices [13]. Theoretically, it has been studied for generating transport in doped [14] and bulk semiconductors through interband [15] and intraband [16] excitations, in graphene and carbon nanotubes [17], and molecular wires [18, 19], among others. The scenario is of interest since, with current laser technology, it can be employed to generate transport on a femtosecond timescale.

An interesting feature of this laser control scenario is that it has both a quantum [11, 12, 14, 20] and a classical [21, 22] manifestation. Further, the two versions of the effect correspond to the same physical phenomenon [7], arising from the nonlinear response of material systems to symmetry breaking radiation fields. In this contribution, we isolate minimum conditions on the driving field and on the system that is being driven that are necessary for the symmetry breaking effect to occur in quantum and classical mechanics. As

shown, the minimum requirements in both cases are identical and, further, the effect can be accounted for in both mechanics through a single symmetry analysis of the equations of motion.

Specifically, in Sec. II we demonstrate that laser rectification can only occur in systems with anharmonic potentials. Subsequently, in Sec. III temporal symmetries of the field that need to be violated to induce transport are isolated. This is done by studying the space-time symmetries of the Liouville equations of motion for laser-driven quantum and classical systems, and by isolating symmetries of the field that rule out any nonzero average currents or dipoles at asymptotic times. In doing so we considerably extend a previous analysis [21] that identified conditions on the field necessary for laser-rectification in classical ergodic systems. It applies to both quantum and classical systems and makes no assumption of ergodicity.

II. CONDITIONS ON THE SYSTEM

Consider first the exact solution for the dynamics of a harmonic oscillator in the presence of an arbitrary space-homogeneous radiation field $E(t)$. The Hamiltonian of the system reads

$$H(x, p) = \frac{p^2}{2m} + \frac{1}{2}m\omega_0^2 x^2 - qE(t)x, \quad (1)$$

where x and p are the position and momentum of the particle of mass m and charge q , and ω_0 is the natural frequency of the oscillator. Symmetry breaking here would correspond to the production of a net dipole moment. In the quantum case, the dynamics of the position $\hat{x}_H(t)$ and momentum operators $\hat{p}_H(t)$ in Heisenberg picture is dictated by the Heisenberg equations of motion

$$\frac{d\hat{x}_H(t)}{dt} = \frac{1}{i\hbar}[\hat{x}_H(t), \hat{H}_H(t)] = \frac{1}{m}\hat{p}_H(t), \quad (2a)$$

$$\frac{d\hat{p}_H(t)}{dt} = \frac{1}{i\hbar}[\hat{p}_H(t), \hat{H}_H(t)] = -m\omega_0^2\hat{x}_H(t) + qE(t), \quad (2b)$$

where \hat{H}_H is the Hamiltonian operator in Heisenberg picture and $[\hat{f}, \hat{H}_H] = \hat{f}\hat{H}_H - \hat{H}_H\hat{f}$ for any operator \hat{f} . In the classical case, the position $x(t)$ and momentum $p(t)$ variables obey

Hamilton's equations

$$\frac{dx(t)}{dt} = \{x(t), H(t)\} = \frac{1}{m}p(t), \quad (3a)$$

$$\frac{dp(t)}{dt} = \{p(t), H(t)\} = -m\omega_0^2 x(t) + qE(t), \quad (3b)$$

where $\{f, H\} = \frac{\partial f}{\partial x} \frac{\partial H}{\partial p} - \frac{\partial f}{\partial p} \frac{\partial H}{\partial x}$ is the Poisson bracket. The difference between Eqs. (2) and (3) is that the former is a differential equation for operators, with operator initial conditions $\hat{x}_H(0) = \hat{x}$ and $\hat{p}_H(0) = \hat{p}$, while the latter is an equation for functions.

These two sets of equations can be solved exactly using Laplace transforms. In fact, for a general external field of the form

$$E(t) = \int_{-\infty}^{\infty} d\omega \epsilon(\omega) e^{i\omega t}, \quad (4)$$

the usual procedure [23] yields:

$$\begin{aligned} \hat{x}_H(t) = \hat{x}_H(0) \cos(\omega_0 t) + \frac{\hat{p}_H(0)}{m\omega_0} \sin(\omega_0 t) \\ + \int_{-\infty}^{\infty} d\omega \frac{q\epsilon(\omega)}{m\omega_0} \frac{i\omega \sin(\omega_0 t) + \omega_0 \cos(\omega_0 t) - \omega_0 e^{i\omega t}}{\omega^2 - \omega_0^2}; \end{aligned} \quad (5)$$

$$\begin{aligned} x(t) = x(0) \cos(\omega_0 t) + \frac{p(0)}{m\omega_0} \sin(\omega_0 t) \\ + \int_{-\infty}^{\infty} d\omega \frac{q\epsilon(\omega)}{m\omega_0} \frac{i\omega \sin(\omega_0 t) + \omega_0 \cos(\omega_0 t) - \omega_0 e^{i\omega t}}{\omega^2 - \omega_0^2}. \end{aligned} \quad (6)$$

The first two terms in Eqs. (5) and (6) describe the field-free evolution of the oscillator, while the third one characterizes the influence of $E(t)$ on the dynamics.

Note that a driven harmonic system can only oscillate at its natural frequency ω_0 and at the frequency of the field ω . That is, there are no frequency components of the dipole that oscillate at multiples or combinations of the frequencies of the field. Hence, if $E(t)$ is unbiased ($\epsilon(0) = 0$) no net dipole can be induced. Thus, we conclude that a necessary requirement for symmetry breaking in quantum and classical mechanics is that the potential of the system is anharmonic. As seen below, the anharmonicities permit the nonlinear response of the system to the incident radiation that mixes the frequencies and harmonics of the field and, for a certain class of radiation sources isolated below, can lead to the generation of a phase-controllable zero-harmonic (dc) component in the response.

III. CONDITIONS ON THE FIELD

We now isolate those temporal symmetries of the field that need to be violated to induce transport in both quantum and classical mechanics. To do so we consider a symmetric one-dimensional system composed of N charged particles coupled to an external field $E(t)$ in the dipole approximation. This is done without loss of generality since the polarization of the field effectively defines an axis along which symmetry breaking can arise. The system's Hamiltonian is then:

$$H = \sum_{j=1}^N \frac{p_j^2}{2m_j} + V(\mathbf{x}) - \sum_{j=1}^N q_j x_j E(t + \alpha \frac{T}{2\pi}), \quad (7)$$

where x_j , p_j , m_j and q_j denote the coordinate, momenta, mass and charge of the j -th particle and $\mathbf{x} \equiv (x_1, x_2, \dots, x_N)$. The systems of interest have a potential $V(\mathbf{x})$ that is invariant under coordinate inversion $V(-\mathbf{x}) = V(\mathbf{x})$, and the driving field $E(t + \alpha \frac{T}{2\pi})$ is an arbitrary time-periodic zero-mean function, with period T and global phase α .

In order to keep a close analogy between the quantum and classical case we frame this analysis in phase space and adopt the Wigner representation of quantum mechanics [24, 25]. In it, the state of the quantum system is described by the Wigner distribution function $\rho_W(\mathbf{x}, \mathbf{p}, t)$, which constitutes a map of the system density matrix $\hat{\rho}$ in the phase space of position \mathbf{x} and momentum \mathbf{p} variables. For N -particle one-dimensional systems it is defined by [25]

$$\rho_W(\mathbf{x}, \mathbf{p}, t) = \left(\frac{1}{2\pi\hbar} \right)^N \int_{-\infty}^{\infty} \dots \int_{-\infty}^{\infty} du_1 \dots du_N e^{\frac{i}{\hbar} \mathbf{p} \cdot \mathbf{u}} \langle \mathbf{x} - \mathbf{u}/2 | \hat{\rho}(t) | \mathbf{x} + \mathbf{u}/2 \rangle, \quad (8)$$

where $|\mathbf{x}\rangle \equiv |x_1\rangle |x_2\rangle \dots |x_N\rangle$ and $\mathbf{p} \cdot \mathbf{u} = \sum_{i=1}^N p_i u_i$. In this way the quantum or classical Liouville evolution can be expressed as

$$\mathcal{D}_\beta \rho_\beta(\mathbf{x}, \mathbf{p}, t) = 0, \quad (9a)$$

where the label β indicates either classical ($\beta = c$) or quantum ($\beta = W$), with $\rho_c(\mathbf{x}, \mathbf{p}, t)$ denoting the classical phase space density. For the Hamiltonian in Eq. (7), the operator \mathcal{D}_β determining the dynamics is given by [25, 26]

$$\mathcal{D}_c = \frac{\partial}{\partial t} - \sum_{j=1}^N \left[-\frac{p_j}{m_j} \frac{\partial}{\partial x_j} + \left(\frac{\partial V(\mathbf{x})}{\partial x_j} - q_j E(t + \alpha \frac{T}{2\pi}) \right) \frac{\partial}{\partial p_j} \right], \quad (9b)$$

$$\mathcal{D}_W = \mathcal{D}_c - \sum_{\substack{\lambda_1, \dots, \lambda_N \\ \lambda_1 + \dots + \lambda_N = 3, 5, \dots}} \frac{(i\hbar/2)^{\lambda_1 + \dots + \lambda_N - 1}}{\lambda_1! \dots \lambda_N!} \frac{\partial^{\lambda_1 + \dots + \lambda_N} V(\mathbf{x})}{\partial x_1^{\lambda_1} \dots \partial x_N^{\lambda_N}} \frac{\partial^{\lambda_1 + \dots + \lambda_N}}{\partial p_1^{\lambda_1} \dots \partial p_N^{\lambda_N}}, \quad (9c)$$

where the last summation in \mathcal{D}_W runs over all positive integer values of $\lambda_1, \dots, \lambda_N$ for which the sum $\lambda_1 + \lambda_2 + \dots + \lambda_N$ is odd and greater than one. In phase space the formal structure of the quantum and classical evolution is remarkably similar [27, 28]. In the limit $\hbar \rightarrow 0$ the second term in Eq. (9c) vanishes and the quantum equation of motion reduces to the classical evolution ($\mathcal{D}_W \rightarrow \mathcal{D}_c$). Note that Eqs. (8) and (9) are fully consistent with the Hamiltonian in Eq. (7). However, when the radiation-matter interaction term in the Hamiltonian goes beyond the dipole approximation both of them need to be modified in order to ensure gauge invariance [26].

In the absence of an external field the equations of motion [Eq. (9)] are invariant under reflection ($\mathbf{x} \rightarrow -\mathbf{x}$, $\mathbf{p} \rightarrow -\mathbf{p}$). Hence, if the system is initially prepared with a given phase-space symmetry this initial symmetry is preserved at all times during the subsequent dynamics. Symmetry breaking is achieved by coupling the system to $E(t)$. However, if $E(t)$ has a zero temporal mean (AC field) then not every $E(t)$ will generate transport. As shown below, by properly lowering the temporal symmetry of $E(t)$ it is possible to induce rectification in the response. Furthermore, the resulting symmetry constraints on $E(t)$ are identical for the classical and quantum case. As will become evident, this is a consequence of the important fact that the quantum corrections in \mathcal{D}_W have the same symmetry properties as $\frac{\partial V(\mathbf{x})}{\partial x_j} \frac{\partial}{\partial p_j}$ under inversion of position and momentum coordinates.

We focus on rectification effects that survive time averaging and that are independent of the global phase α of the laser beam. Typically, symmetry breaking effects that depend on α are difficult to control (although not impossible [29]) since this requires an experimental setup that both locks the absolute phase of the laser and has control over the center of mass motion with respect to the laboratory frame. Hence, the quantities of interest are the mean position and momentum averaged over time and over α :

$$\langle \overline{\overline{\mathbf{x}}} \rangle_\beta = \lim_{\tau \rightarrow \infty} \int_{-\tau/2}^{\tau/2} \frac{dt}{\tau} \int_0^{2\pi} \frac{d\alpha}{2\pi} \text{Tr}(\mathbf{x} \rho_\beta(\mathbf{x}, \mathbf{p}, t)); \quad (10a)$$

$$\langle \overline{\overline{\mathbf{p}}} \rangle_\beta = \lim_{\tau \rightarrow \infty} \int_{-\tau/2}^{\tau/2} \frac{dt}{\tau} \int_0^{2\pi} \frac{d\alpha}{2\pi} \text{Tr}(\mathbf{p} \rho_\beta(\mathbf{x}, \mathbf{p}, t)); \quad (10b)$$

where the double overbar indicates this kind of averaging. Here the notation $\langle \dots \rangle_\beta$ denotes the classical ensemble average ($\beta = c$) or quantum expectation value ($\beta = W$), and the trace is an integration over the $2N$ -dimensional phase-space (\mathbf{x}, \mathbf{p}) . When the symmetry of the system is not broken, both $\langle \overline{\overline{\mathbf{x}}} \rangle_\beta$ and $\langle \overline{\overline{\mathbf{p}}} \rangle_\beta$ are zero. Below we determine symmetries of the

field and of the initial condition that guarantee that this is indeed the case. When these symmetries are violated a net dipole or current is expected to appear.

The fact that we are only interested in α -independent properties eliminates the necessity to invoke ergodicity in the analysis. The average over α is sufficient to obviate any initial-time preparation effects, which is the main role of the ergodicity assumption in the purely classical analysis of Ref. [21].

We now tabulate the symmetries of the field that will be relevant for our purposes. The field may change sign every half a period T ,

$$E(t + T/2) = -E(t); \quad (11a)$$

or be symmetric or antisymmetric with respect to some time t'

$$E(t - t') = +E(-(t - t')); \quad (11b)$$

$$E(t - t') = -E(-(t - t')). \quad (11c)$$

Each of the symmetries in Eq. (11) leads to a transformation that leaves the equations of motion invariant while changing the sign of the position and/or momentum variables. They are identical for the quantum and classical case. For example, if $E(t)$ satisfies Eq. (11a), then \mathcal{D}_β is invariant under \mathcal{T}_1 defined as:

$$\mathcal{T}_1 : \quad t \rightarrow t + T/2; \quad \mathbf{x} \rightarrow -\mathbf{x}; \quad \mathbf{p} \rightarrow -\mathbf{p}; \quad (12a)$$

where we have taken into account that under inversion of position and momenta, $\frac{\partial^{\lambda_1 + \dots + \lambda_N}}{\partial x_1^{\lambda_1} \dots \partial x_N^{\lambda_N}} \rightarrow -\frac{\partial^{\lambda_1 + \dots + \lambda_N}}{\partial x_1^{\lambda_1} \dots \partial x_N^{\lambda_N}}$ and $\frac{\partial^{\lambda_1 + \dots + \lambda_N}}{\partial p_1^{\lambda_1} \dots \partial p_N^{\lambda_N}} \rightarrow -\frac{\partial^{\lambda_1 + \dots + \lambda_N}}{\partial p_1^{\lambda_1} \dots \partial p_N^{\lambda_N}}$ since $\lambda_1 + \lambda_2 + \dots + \lambda_N$ in Eq. (9c) is odd. Similarly, if $E(t)$ satisfies Eq. (11b) [or Eq. (11c)] then \mathcal{D}_β is invariant under \mathcal{T}_2 [or \mathcal{T}_3], where

$$\mathcal{T}_2 : \quad t - t' \rightarrow -(t - t'); \quad \mathbf{x} \rightarrow \mathbf{x}; \quad \mathbf{p} \rightarrow -\mathbf{p}; \quad (12b)$$

$$\mathcal{T}_3 : \quad t - t' \rightarrow -(t - t'); \quad \mathbf{x} \rightarrow -\mathbf{x}; \quad \mathbf{p} \rightarrow \mathbf{p}. \quad (12c)$$

Other temporal symmetries of the field exist but play no role in this analysis since they do not lead to invariance transformations that change the sign of the position and/or momentum variables.

Now, given a solution to Eq. (9), $\rho_\beta(\mathbf{x}, \mathbf{p}, t)$, if \mathcal{D}_β is invariant under \mathcal{T}_α one can generate another solution to the same equation by applying \mathcal{T}_α to $\rho_\beta(\mathbf{x}, \mathbf{p}, t)$. The new solutions

$\rho_\beta^{(\alpha)}(\mathbf{x}, \mathbf{p}, t) = \mathcal{T}_\alpha \rho_\beta(\mathbf{x}, \mathbf{p}, t)$ generated by the invariance transformations in Eq. (12) are:

$$\rho_\beta^{(1)}(\mathbf{x}, \mathbf{p}, t) = \mathcal{T}_1 \rho_\beta(\mathbf{x}, \mathbf{p}, t) = \rho_\beta(-\mathbf{x}, -\mathbf{p}, t + T/2); \quad (13a)$$

$$\rho_\beta^{(2)}(\mathbf{x}, \mathbf{p}, t) = \mathcal{T}_2 \rho_\beta(\mathbf{x}, \mathbf{p}, t - t') = \rho_\beta(\mathbf{x}, -\mathbf{p}, -(t - t')); \quad (13b)$$

$$\rho_\beta^{(3)}(\mathbf{x}, \mathbf{p}, t) = \mathcal{T}_3 \rho_\beta(\mathbf{x}, \mathbf{p}, t - t') = \rho_\beta(-\mathbf{x}, \mathbf{p}, -(t - t')). \quad (13c)$$

Further, if the original solution $\rho_\beta(\mathbf{x}, \mathbf{p}, t)$ predicts an average position $\langle \bar{\mathbf{x}} \rangle_\beta$ and momenta $\langle \bar{\mathbf{p}} \rangle_\beta$, the transformed solutions $\rho_\beta^{(\alpha)}(\mathbf{x}, \mathbf{p}, t)$ will predict a mean position $\langle \bar{\mathbf{x}} \rangle_\beta^{(\alpha)}$ and/or momenta $\langle \bar{\mathbf{p}} \rangle_\beta^{(\alpha)}$ that has the same magnitude but is opposite in sign:

$$\langle \bar{\mathbf{x}} \rangle_\beta^{(1)} = -\langle \bar{\mathbf{x}} \rangle_\beta; \quad \langle \bar{\mathbf{p}} \rangle_\beta^{(1)} = -\langle \bar{\mathbf{p}} \rangle_\beta; \quad (14a)$$

$$\langle \bar{\mathbf{x}} \rangle_\beta^{(2)} = +\langle \bar{\mathbf{x}} \rangle_\beta; \quad \langle \bar{\mathbf{p}} \rangle_\beta^{(2)} = -\langle \bar{\mathbf{p}} \rangle_\beta; \quad (14b)$$

$$\langle \bar{\mathbf{x}} \rangle_\beta^{(3)} = -\langle \bar{\mathbf{x}} \rangle_\beta; \quad \langle \bar{\mathbf{p}} \rangle_\beta^{(3)} = +\langle \bar{\mathbf{p}} \rangle_\beta. \quad (14c)$$

The argument is completed by showing that the average position and momenta predicted by $\rho_\beta(\mathbf{x}, \mathbf{p}, t)$ and $\rho_\beta^{(\alpha)}(\mathbf{x}, \mathbf{p}, t)$ coincide. If this is the case, it follows from Eq. (14) that symmetry breaking cannot occur. For this we exploit the possible symmetries of the initial state:

$$\rho_\beta(\mathbf{x}, \mathbf{p}, t_0) = \rho_\beta(-\mathbf{x}, -\mathbf{p}, t_0); \quad (15a)$$

$$\rho_\beta(\mathbf{x}, \mathbf{p}, t_0) = \rho_\beta(\mathbf{x}, -\mathbf{p}, t_0); \quad (15b)$$

$$\rho_\beta(\mathbf{x}, \mathbf{p}, t_0) = \rho_\beta(-\mathbf{x}, \mathbf{p}, t_0). \quad (15c)$$

The first one corresponds to a state with zero mean position and momenta, while the second and third describe an initial state with either zero mean momenta or zero mean position, respectively.

Consider the case in which the equations of motion are \mathcal{T}_1 invariant. The distributions $\rho_\beta(\mathbf{x}, \mathbf{p}, t)$ and $\rho_\beta^{(1)}(\mathbf{x}, \mathbf{p}, t)$ satisfy the same equation of motion but do not, in general, coincide. However, if the initial condition for the original solution $\rho_\beta(\mathbf{x}, \mathbf{p}, t_0)$ is invariant under reflection [Eq. (15a)] then

$$\rho_\beta^{(1)}(\mathbf{x}, \mathbf{p}, t_0 - T/2) = \rho_\beta(-\mathbf{x}, -\mathbf{p}, t_0) = \rho_\beta(\mathbf{x}, \mathbf{p}, t_0) = \rho_\beta^{(1)}(-\mathbf{x}, -\mathbf{p}, t_0 - T/2). \quad (16)$$

That is, the original and transformed solutions start from the same initial distribution but at initial time they experience a different value for the global phase of the field, $E(t_0 + \alpha \frac{T}{2\pi})$ and

$E(t_0 + (\alpha - \pi)\frac{T}{2\pi}) = -E(t_0 + \alpha\frac{T}{2\pi})$ respectively. Since the averages in Eq. (10) are independent of α , they coincide for the two solutions. Hence, no rectification can be induced when the field satisfies Eq. (11a) and the initial condition satisfies Eq. (15a).

The argument for the two other cases is very similar. If the field satisfies Eq. (11b) [or Eq. (11c)], the equation of motion is \mathcal{T}_2 (or \mathcal{T}_3) invariant. Even when the original $\rho_\beta(\mathbf{x}, \mathbf{p}, t)$ and transformed $\rho_\beta^{(2)}(\mathbf{x}, \mathbf{p}, t)$ [or $\rho_\beta^{(3)}(\mathbf{x}, \mathbf{p}, t)$] solutions obey the same equation of motion, they do not need to coincide. However, if the initial condition of the original solution satisfies the symmetry in Eq. (15b) [or Eq. (15c)], then

$$\rho_\beta^{(2)}(\mathbf{x}, \mathbf{p}, -t_0 + t') = \rho_\beta(\mathbf{x}, -\mathbf{p}, t_0) = \rho_\beta(\mathbf{x}, \mathbf{p}, t_0) = \rho_\beta^{(2)}(\mathbf{x}, -\mathbf{p}, -t_0 + t') \quad (17)$$

$$\rho_\beta^{(3)}(\mathbf{x}, \mathbf{p}, -t_0 + t') = \rho_\beta(-\mathbf{x}, \mathbf{p}, t_0) = \rho_\beta(\mathbf{x}, \mathbf{p}, t_0) = \rho_\beta^{(3)}(-\mathbf{x}, \mathbf{p}, -t_0 + t') \quad (18)$$

The transformed solution has the same initial condition as the original one but as we had prepared the system a time $2t_0 - t'$ before. The difference between the two solutions is that they experience a different global laser phase at preparation time. Since we are not interested in effects that depend on the global laser phase, the mean position and momenta in Eq. (10) for the original and transformed solution need to coincide. However, from Eq. (14b) [or Eq. (14c)] we conclude that this can only happen if $\langle \bar{\mathbf{p}} \rangle_\beta = 0$ [or $\langle \bar{\mathbf{x}} \rangle_\beta = 0$].

In summary, for spatially symmetric *classical or quantum* systems initially prepared in a symmetric state that satisfies Eq. (15), net transport using time-periodic external fields with zero temporal mean can only be generated if the field violates the temporal symmetries in Eq. (11). Further, any symmetry breaking effect that may be achieved with a field that satisfies Eq. (11) is necessarily due to an effect that depends on the global phase of the laser (cf. Ref. [29]).

It is natural to ask what kind of fields have sufficiently low temporal symmetry to induce net transport. Monochromatic sources satisfy all the symmetries in Eq. (11) and, as a consequence, cannot be used to induce symmetry breaking. However, by adding a second frequency component to a monochromatic source it is possible to lower the symmetry of the field and induce symmetry breaking. For instance, a field like

$$E(t) = \epsilon_{n\omega} \cos(n\omega t + \phi_{n\omega}) + \epsilon_{m\omega} \cos(m\omega t + \phi_{m\omega}), \quad (19)$$

where n and m are coprime integers so that $E(t)$ has a period $T = 2\pi/\omega$, satisfies Eq. (11) only under special conditions. It satisfies (11a) only if n and m are odd. Thus, a field with

$n = 3$ and $m = 1$, like the one used in the 1 vs. 3 photon control scenario [1], will not be symmetry breaking. However, a field with $n = 2$ and $m = 1$, like the one employed in the 1 vs. 2 scenario, does not satisfy Eq. (11) and is expected to induce net dipoles and currents. These dipoles and currents are phase-controllable since, by varying the relative phase between the two components of the beam, the $\omega + 2\omega$ field may satisfy Eq. (11b) or (11c) and thus rule out the possibility of inducing currents or dipoles, respectively. Explicitly, when $\phi_{2\omega} - 2\phi_{\omega} = 0, \pm\pi, \pm2\pi, \dots$ an $\omega + 2\omega$ field satisfies Eq. (11b) and zero currents are expected. Similarly, when $\phi_{2\omega} - 2\phi_{\omega} = \pm\frac{\pi}{2}, \pm\frac{3\pi}{2}, \dots$ it fulfills symmetry (11c) and no dipoles can be induced. For all other choices of the phases an $\omega + 2\omega$ field is expected to induce symmetry breaking.

IV. CONCLUSIONS

In conclusion, we have shown that the minimum conditions for the generation of phase controllable asymmetry in spatially symmetric quantum and classical systems using time-periodic external fields with zero temporal mean are identical: anharmonicities in the system's potential are required as is a driving field that violates the temporal symmetries in Eq. (11). These conditions refer to symmetry breaking effects that do not rely on the control of the absolute phase of the field. The derived results provide necessary conditions for the generation of asymmetry, applicable to all systems. Additional sufficient conditions may be required, but these depend upon the specific system under consideration.

Further, we have shown that both quantum and classical versions of the symmetry breaking effect can be accounted for through a single space-time symmetry analysis of the equations of motion. The anharmonicities in the potential permit the nonlinear response of the system to the incident radiation that, through harmonic mixing, and for fields that violate Eq. (11), can lead to the generation of a phase-controllable DC component in the photoinduced dipoles or currents.

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- [1] M. Shapiro and P. Brumer, *Principles of the Quantum Control of Molecular Processes* (John Wiley & Sons, New York, 2003).
- [2] S. A. Rice and M. Zhao, *Optical Control of Molecular Dynamics* (John Wiley & Sons, New York, 2000).
- [3] K. Bergmann, H. Theuer, and B. W. Shore, Rev. Mod. Phys. **70**, 1003 (1998).
- [4] S. A. Rice, Nature **409**, 422 (2001).
- [5] M. Dantus and V. V. Lozovoy, Chem. Rev. **104**, 1813 (2004).
- [6] P. Nuernberger, G. Vogt, T. Brixner, and G. Gerber, Phys. Chem. Chem. Phys. **9**, 2470 (2007).
- [7] I. Franco and P. Brumer, Phys. Rev. Lett. **97**, 040402 (2006).
- [8] Y. Y. Yin, C. Chen, D. S. Elliott, and A. V. Smith, Phys. Rev. Lett. **69**, 2353 (1992).
- [9] B. Sheehy, B. Walker, and L. F. DiMauro, Phys. Rev. Lett. **74**, 4799 (1995).
- [10] E. Charron, A. Giusti-Suzor, and F. H. Mies, Phys. Rev. Lett. **75**, 2815 (1995).
- [11] E. Dupont, P. B. Corkum, H. C. Liu, M. Buchanan, and Z. R. Wasilewski, Phys. Rev. Lett. **74**, 3596 (1995).
- [12] A. Haché, Y. Kostoulas, R. Atanasov, J. L. P. Hughes, J. E. Sipe, and H. M. van Driel, Phys. Rev. Lett. **78**, 306 (1997).
- [13] M. Schiavoni, L. Sanchez-Palencia, F. Renzoni, and G. Grynberg, Phys. Rev. Lett. **90**, 094101 (2003).
- [14] G. Kurizki, M. Shapiro, and P. Brumer, Phys. Rev. B **39**, 3435 (1989).
- [15] R. Atanasov, A. Haché, J. L. P. Hughes, H. M. van Driel, and J. E. Sipe, Phys. Rev. Lett. **76**, 1703 (1996).
- [16] K. A. Pronin and A. D. Bandrauk, Phys. Rev. B **69**, 195308 (2004).
- [17] E. J. Mele, P. Král, and D. Tománek, Phys. Rev. B **61**, 7669 (2000).
- [18] I. Franco, M. Shapiro, and P. Brumer, Phys. Rev. Lett. **99**, 126802 (2007).
- [19] J. Lehmann, S. Kohler, V. May, and P. Hanggi, J. Chem. Phys. **121**, 2278 (2004).

- [20] I. Goychuk and P. Hänggi, *Europhys. Lett.* **43**, 503 (1998).
- [21] S. Flach, O. Yevtushenko, and Y. Zolotaryuk, *Phys. Rev. Lett.* **84**, 2358 (2000).
- [22] S. Denisov, S. Flach, A. A. Ovchinnikov, O. Yevtushenko, and Y. Zolotaryuk, *Phys. Rev. E* **66**, 041104 (2002).
- [23] G. B. Arfken and H. J. Weber, *Mathematical Methods for Physicists* (Harcourt Academic Press, U.S.A., 2001), 5th ed.
- [24] V. I. Tatarskii, *Sov. Phys. Usp.* **26**, 311 (1983).
- [25] M. Hillery, R. F. O’Connell, M. O. Scully, and E. P. Wigner, *Phys. Rep.* **106**, 121 (1984).
- [26] O. T. Serimaa, J. Javanainen, and S. Varró, *Phys. Rev. A* **33**, 2913 (1986).
- [27] J. Wilkie and P. Brumer, *Phys. Rev. A* **55**, 27 (1997).
- [28] J. Wilkie and P. Brumer, *Phys. Rev. A* **55**, 43 (1997).
- [29] M. F. Kling, C. Siedschlag, A. J. Verhoef, J. I. Khan, M. Schultze, T. Uphues, Y. Ni, M. Uiberacker, M. Drescher, F. Krausz, et al., *Science* **312**, 246 (2006).